Removal of pharmaceuticals in sewage treatment plants: A model generalisation to international data

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Removal of pharmaceuticals in sewage treatment plants: A model generalisation to international data

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Abstract: The removal efficiency is used in literature as key indicator of the fate of pharmaceuticals in wastewater treatment plants. In this study, we used the recently developed Activated Sludge Modelling framework for Xenobiotics (ASM-X) to identify and evaluate the mechanisms affecting the removal efficiency in biological treatment systems. Model predictions for sulfamethoxazole, ciprofloxacin and tetracycline were generalized with published data derived from a critical literature selection. The biological removal efficiency was significantly underestimated when assessed based only on parent chemical fractions, showing the significance of retransformation processes. We also demonstrated that a dynamic evaluation of the removal of pharmaceuticals could lead to a more detailed assessment of the potential environmental risk.

Keywords: Micropollutants removal; retransformation of pharmaceuticals; ASM-X generalization

Introduction

Biological wastewater treatment plants (WWTPs) have a crucial role in the mitigation of the environmental risk posed by the release of pharmaceuticals. A number of factors are known to influence the removal of pharmaceuticals. Among others, we note: (i) dynamics in the influent loads (related to e.g., administration patterns) and in the operation of WWTPs; (ii) the sludge retention time (SRT) of biological systems, possibly inducing an increase in the metabolic capabilities of biomass; and (iii) the occurrence of retransformation processes (e.g., deconjugation of human metabolites) in upstream sewers and in bioreactors. With regard to the latter, the concentration of human metabolites that can retransform to the parent form was found to be greater than the parent concentration itself (Göbel et al., 2005). The assessment of the actual removal processes can therefore be misleading, if the removal efficiency is estimated by using data only on parent chemicals—as usually occurring in literature.

An Activated Sludge Modelling framework for Xenobiotics (ASM-X) was recently tested in the fate prediction of pharmaceuticals in an activated sludge WWTP (Plósz et al., 2010, 2012). Based on these predictions, we generalized the removal of three pharmaceuticals (sulfamethoxazole, ciprofloxacin, tetracycline) with worldwide literature data. The objectives of this generalization were: (i) to validate ASM-X predictions with literature data; and (ii) to assess the effects of the mentioned influencing factors on the full-scale removal of the substances. Additionally, we investigated the effect of effluent dynamics on the risk prediction in recipient bodies.

Material and Methods

Based on the ASM-X predictions presented by Plósz et al. (2010), average removal efficiencies [%] of sulfamethoxazole, ciprofloxacin and tetracycline in Bekkelaget WWTP (Oslo, Norway) were estimated and plotted as a function of the influent load of the substances [mg h^{-1} $1000PE^{-1}$]. Distinct removal efficiencies were calculated considering i) only the parent compound fraction and ii) both the parent and the

retransformable fractions. International data on the removal of pharmaceuticals, resulting from WWTP measuring campaigns, were used for the generalization. A number of criteria were defined for the literature selection, e.g. the use of suitable wastewater sampling techniques. An environmental risk assessment of the chemicals was performed by calculating predicted environmental concentrations (PECs) from the effluent concentrations reported by Plósz et al. (2010).

Results and Conclusions

For a meaningful comparison of removal efficiencies at the influent loadings reported in literature, scenario simulations with ASM-X were considered. Model predictions with 5-fold increased influent loading were found to reproduce the efficiencies reported by Göbel et al. (2005, 2007) (Fig. 1). A significant underestimation error occurred when only the parent fraction was considered, as compared to when N4-acetyl-sulfamethoxazole (a major metabolite) was accounted. The comparable elimination performances of the WWTPs assessed could be associated to similar operational conditions in the WWTPs and impacts of in-sewer retransformation on influent characteristics. Removal efficiencies reported by Radjenovic et al. (2009)—based on only parent removal—were higher than predicted ones both for activated sludge and membrane bioreactor, suggesting that in-sewer retransformation could have overcome the effect of SRT (> 60 d) increase in the latter bioreactor type. Furthermore, the comparably higher efficiencies reported by Yang et al. (2011) were possibly related to the employment of membrane filtration in the monitored WWTP.

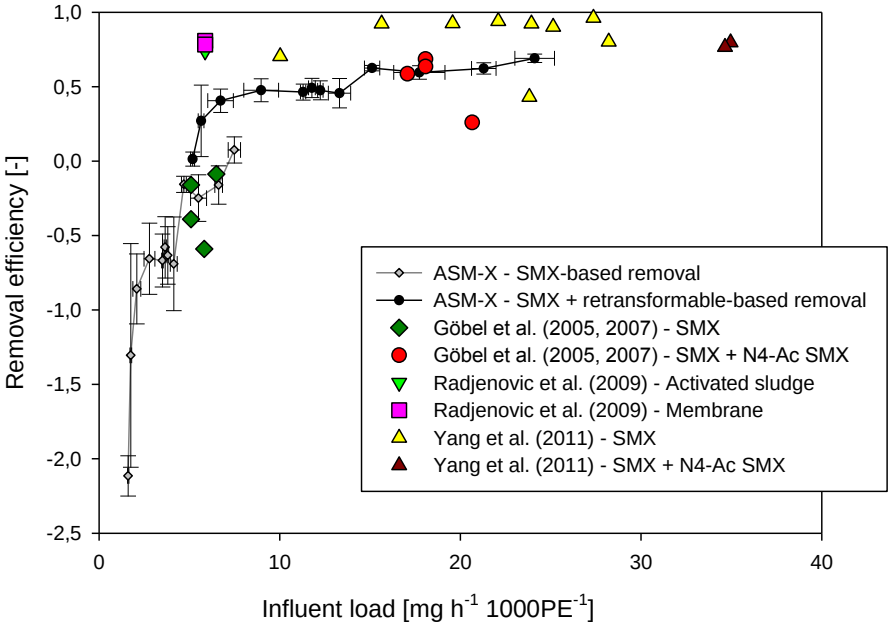

Figure 1 Generalization of predicted removal of sulfamethoxazole (SMX) through ASM-X with literature data. ASM-X predictions of removal efficiency accounted for only parent SMX (grey diamonds) and for parent and retransformable fractions of SMX (black circles). Error bars refer to standard deviations in influent loadings and removal efficiencies. Removal efficiencies from Göbel et al. (2005, 2007) were calculated based as average of published data on SMX and N4-acetyl-sulfamethoxazole (N4-Ac SMX).

Results from the preliminary risk assessment showed the potential risk associated to sulfamethoxazole (Fig. 2a) and tetracycline (Fig. 2b) in the recipient water bodies of Bekkelaget. A low additional risk potential was related to the retransformable fraction

of sulfamethoxazole, as a result of its comparably high retransformation rate in activated sludge (Plósz et al., 2010). On the other hand, the retransformable fraction of tetracycline was found to substantially impact the PEC value of the substance (up to 130% increase as compared to parent-based PEC).

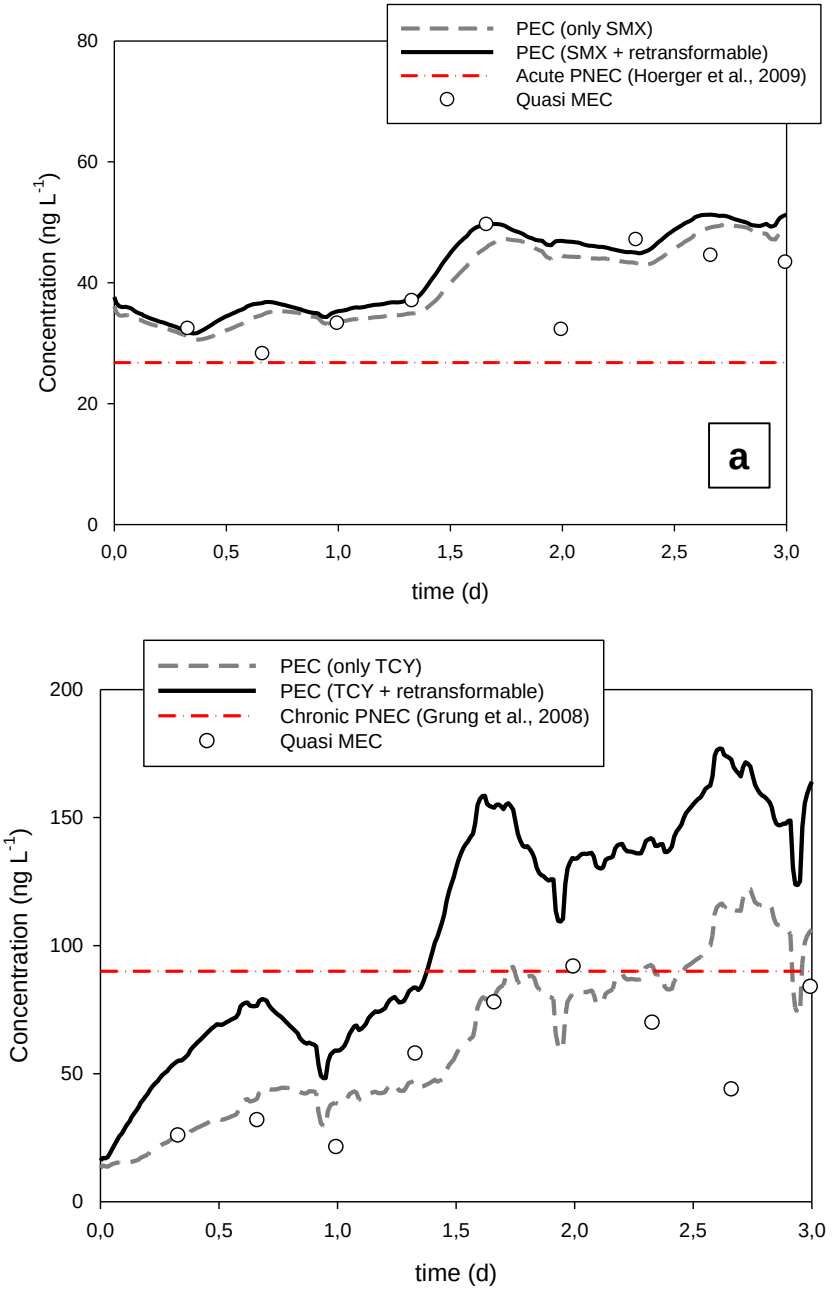

Figure 2 PEC values of sulfamethoxazole (SMX-a) and tetracycline (TCY-b), calculated from ASM-X predictions. We distinguished between PECs accounting for only parent (grey dashed line) and parent plus retransformable (black line). For the purpose of risk assessment, PECs were compared with acute and chronic predicted non-effect concentrations (PNEC). Quasi MECs (measured environmental concentrations) identify effluent measured environmental concentrations divided by a dilution factor.

These results suggest that a comprehensive evaluation of the removal of pharmaceuticals in biological WWTPs is required for a more detailed assessment of the risk derived by their environmental release.

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